

RETENTION OF RE IN METAKAOLIN BASED GEOPOLYMER IN THE PRESENCE OF AN ORGANIC REDUCTANT – AN EXPERIMENTAL STUDY

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Key Words: geopolymer, metakaolin, rhenium, oxidation, leaching

The goal of this research was to examine the change in redox sensitive contaminant retention and leaching behavior from a model geopolymer system, during oxidation and carbonation, in the presence of an organic reductant. The behavior of redox sensitive elements under variable environmental conditions is specifically important for understanding the leaching of radionuclides retained in geopolymeric materials (e.g. Tc) that are soluble when oxidized and insoluble under reducing conditions.

An alkali-activated metakaolin geopolymer with high silica/alumina ratio was selected as a model material. Rhenium was selected as the model redox sensitive element, which exhibits a large valence (-3 to +7) and solubility range. Rhenium was introduced in its most oxidized form (VII) as Re_2S_7 . Ascorbic acid was used as a reducing agent during geopolymer casting due to its high solubility and homogeneous distribution within the matrix. Cylindrical monoliths were cured for 90 days under an inert atmosphere (N_2) and then aged for 60 days under 98% N_2 + 2% CO_2 or CO_2 deficient air, at a constant relative humidity (68%). The samples were then subjected to EPA 1313 pH dependent leaching test and EPA 1315 monolith leaching test. Leachates were analyzed using DOC and ICP techniques. XRD and SEM – EDS imaging and analyses were used to characterize the solids

Results show that Re has precipitated as ReS_2 (IV) in the matrix. The pH dependent leaching test show that Re was released homogeneously throughout most of the pH range (3-12). However, the retention of the material aged under CO_2 deficient air was three times lower, indicating that the main effect on leaching from this material is of the oxidation process. The natural pH values were 11.3 and 12 for the material aged under 2% CO_2 and for material aged under CO_2 deficient air respectively, indicating minor carbonation in the former, however, no carbonate minerals were detected in the matrix.

Monolith leaching results show that the geopolymers aged under CO_2 deficient air have retained significantly less Re (0.24% leaching) relative to the sample aged under 2% CO_2 (0.07wt % leaching). $-\log(\text{diffusivity})$ values were in the order of 15 and 16 for CO_2 deficient air and 2% CO_2 respectively. There is a clear positive correlation between the amount of Re and DOC released from the samples, where their amounts are significantly larger for the samples aged under CO_2 deficient air, indicating that the main Re-release mechanism from these samples is related to chelation to organic species under oxidizing conditions.